

What is claimed is:

1. A double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state from a ground state, a light source for an erase light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher excited state from the first electronic excited state and overlap means for partially overlapping irradiating areas of the pump light and the erase light with each other, wherein an emission area upon deexcitation of the sample molecule from the first electronic excited state to the ground state is partially inhibited by irradiating the pump light and the erase light through the overlap means, said double-resonance-absorption microscope characterized in that there is provided, on an optical path of the erase light, a spatial filter which has a condenser lens and a collimate lens and a pinhole therebetween and performs condensing of the erase light onto the pinhole by the condenser lens and collimating of the erase light passed through the pinhole into a parallel beam by the collimate lens.

2. The double-resonance-absorption microscope according to claim 1, wherein a radius of the pinhole satisfies a condition expressed by

$$\text{either } a \leq \frac{f\lambda_2}{8nh} \text{ or } a \leq \frac{\lambda_2}{8nNA}.$$

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3. The double-resonance-absorption microscope according to claim 1 or 2, further comprising a phase modulation element which gives, to the erase light passed through the spatial filter, a phase difference of  $\pi$  around an optical axis of the erase light as the optical axis being a center.

4. The double-resonance-absorption microscope according to claim 3, wherein the phase modulation element gives, to the erase light passed through a means for a limitation of a beam area, a phase difference of  $\pi$  around an optical axis of the erase light as the optical axis being a center.

5. The double-resonance-absorption microscope according to claim 4, wherein the phase modulation element comprises a parallel substrate which is transparent and optical flat to the erase light and a optical thin film, evaporated on the parallel substrate, which has a thickness distribution capable of giving the phase difference of  $\pi$  to the erase light around the optical axis thereof as the optical axis being the center.

6. The double-resonance-absorption microscope according to claim 4, wherein the phase modulation element comprises a parallel substrate which is transparent and optical flat to the erase light and also is performed a etching capable of giving the phase difference of  $\pi$  to the erase light around the optical axis thereof as the optical axis being the center.

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7. The double-resonance-absorption microscope according to any one of claims 1 to 6, wherein a sample is dyed with a

~~fluorescence labeler molecule having at least three electronic state including a ground state and the sample molecule is the fluorescence labeler molecule.~~

8. A double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state from a ground state, a light source for an erase light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher excited state from the first electronic excited state and overlap means for partially overlapping irradiating areas of the pump light and the erase light with each other, wherein an emission area upon deexcitation of the sample molecule to the ground state from the first electronic excited state is partially inhibited by irradiating the pump light and the erase light through the overlap means, said double-resonance-absorption microscope characterized in that a coherent light is provided as the light source for the erase light and the erase light by the coherent light source has a phase distortion in a beam plane less than  $\lambda_2/2$ .

9. The double-resonance-absorption microscope according to claim 8, wherein a sample is dyed with a fluorescence labeler molecule having at least three electronic state including a ground state and the sample molecule is the fluorescence labeler molecule.

~~10. The double-resonance-absorption microscope according to~~

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claim 8 or 9, further comprising beam area limitation means for extracting only an area having the beam distortion in the beam plane less than  $\lambda_2/2$  from the erase light by the coherent light source, wherein the erase light is made to have the phase distortion in the beam plane less than  $\lambda_2/2$  by the beam area limitation means.

11. The double-resonance-absorption microscope according to claim 10, further comprising a phase modulation element which gives a spatial phase distribution in a beam cross-section to the erase light passed through the beam area limitation means.

12. The double-resonance-absorption microscope according to claim 11, wherein the phase modulation element gives, to the erase light passed through the beam area limitation means, a phase difference of  $\pi$  around an optical axis of the erase light as the optical axis being a center.

13. The double-resonance-absorption microscope according to claim 11, wherein the phase modulation element gives, to the erase light passed through the beam area limitation means, the spatial phase distribution on a concentric circle in a cross-section of the erase light.

14. The double-resonance-absorption microscope according to claim 11, wherein the phase modulation element gives, to the erase light passed through the beam area limitation means, the spatial phase distribution having a phase difference of  $\pi$  between a circle area and a ring area outside of the circle area in a

cross-section of the erase light.

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15. The double-resonance-absorption microscope according to any one of claims 11 to 14, wherein the phase modulation element comprises a parallel substrate which is optical flat to the erase light and an optical thin film, evaporated on the parallel substrate, which has a prescribed thickness distribution.

16. The double-resonance-absorption microscope according to any one of claims 11 to 14, wherein the phase modulation element comprises a parallel substrate which is optical flat to the erase light and also is performed a etching.

17. The double-resonance-absorption microscope according to any one of claims 8 to 16, further comprising fluorescence detection means having a photoelectron conversion plane, a microchannel plate, a phosphor screen, an optical fiber coupler, and a CCD detector.

18. The double-resonance-absorption microscope according to claim 17, wherein the fluorescence detection means has, in front of the photoelectron conversion plane, a light separation element or a wavelength dispersion element which removes either or both of the pump light and the erase light from a fluorescence signal.

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19. The double-resonance-absorption microscope according to claim 17 or 18, further comprising a slit or a pinhole on an optical axis between a sample plane and the fluorescence detection means.

20. The double-resonance-absorption microscope according to

claim 19, wherein the slit or the pinhole is provided immediately in front of the photoelectron conversion plane.

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21. The double-resonance-absorption microscope according to any one of claims 17 to 20, further comprising time control means for controlling the time to apply a voltage to an electrode of each of the photoelectron converter, the microchannel plate and the phosphor screen.

22. The double-resonance-absorption microscope according to 21, wherein the time control means controls the time to apply the voltage by a switching and a switching response time thereof is shorter than a fluorescence time of the sample molecule.

23. The double-resonance-absorption microscope according to 21, wherein the time control means controls the time to apply the voltage so that the time becomes longer than a fluorescence time of the sample molecule.

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24. The double-resonance-absorption microscope according to any one of claims 8 to 16, further comprising a light separation element or a wavelength dispersion element which removes either or both of the pump light and the erase light from a fluorescence signal.

25. The double-resonance-absorption microscope according to any one of claims 6 to 24, wherein a pulse width of the erase light is longer than that of the pump light and also an irradiating time of the pump light completely overlaps with that of the erase light.

26. The double-resonance-absorption microscope according to claim 25, further comprising pulse width control means for widening a pulse width of the erase light so that it becomes longer than that of the pump light.

27. The double-resonance-absorption microscope according to claim 26, further comprising a pulse stretcher optical system, as the pulse width control means, which has a half mirror for a light separation and a reflection optical system for forming a loop optical path including the half mirror thereon.

28. The double-resonance-absorption microscope according to claim 25, further comprising irradiating timing control means for controlling the time when the pump light and the erase light reach to the sample so that the irradiation time of the pump light completely overlaps with that of the erase light.

29. The double-resonance-absorption microscope according to claim 28, wherein the irradiation timing means controls the time when the pump light and the erase light reach to the sample by controlling an optical path difference of the pump light and the erase light.

30. The double-resonance-absorption microscope according to claim 28, wherein the light source for the pump light and the light source for the erase light are pulse lasers independent from each other and the irradiation time control means controls the time when the pump light and the erase light reach to the sample by controlling a Q-switch of each of the pulse laser.

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31. The double-resonance-absorption microscope according to any one of claims 8 to 30, wherein there is used a standard sample having a substrate transparent to the pump light and the erase light and a molecule uniformly applied to the substrate, capable of being excited by either of the pump light and the erase light, and wherein optical axes of the pump light and the erase light are controlled so that an area and a luminance of a light emission becomes minimum and maximum, respectively, said light emission occurs from the molecule when the pump light and the erase light are simultaneously applied to the standard sample.

32. The double-resonance-absorption microscope according to any one of claims 8 to 31, further comprising a mechanism for scanning the pump light and the erase light or scanning the sample at least with a nanometer order.

33. The double-resonance-absorption microscope according to claim 32, having a sample stage mechanism capable to move two-dimensionally by an electromagnetic motor and a piezoelectric element as the mechanism for scanning of the sample.

34. The double-resonance-absorption according to claim 32, having a swaying mirror mechanism as the mechanism for scanning the pump light and the erase light.

35. A solid dye laser comprising:

a solid laser medium where a dye molecule having at least more than two quantum levels is dispersed; and



a short pulse laser which excites the solid laser medium.

36. The solid dye laser according to claim 35, wherein the solid laser medium is made by applying a sol-gel method to an inorganic solution and an organic solution where the dye molecule is dispersed.

37. The solid dye laser according to claim 35, wherein the solid laser medium is made by applying a deliquescent heating method to a solid organic material where the dye molecule is dispersed.

38. The solid dye laser according to claim 35, wherein the dye molecule is a molecule having a double bond or a benzene ring.

39. The solid dye laser according to claim 38, wherein the dye molecule is any one of

2,2"-Dimethyl-p-terphenyl:

P-terphenyl (PTP):

3,3',2",3"-Tetramethyl-P-quaterphenyl:

2,2'''-Dimethyl-P-quaterphenyl:

2-Methyl-5-t-butyl-p-quaterphenyl:

2-(4-Biphenyllyl)-5-(4-t-butylphenyl)-1,3,4-oxiazol (BPBD-365):

2-(4-Biphenyllyl)-phenyl-1,3,4-oxadiazol:

2,5,2''''5, ''''-Tetramethyl-p-quinquephenyl:

3,5,3''''5, ''''-Tetra-t-butyl-p-quinquephenyl:

2,5-Diphenyloxazol:

2,5-Diphenylfuran:

PQP(p-Quanterphenyl):

2,5-Bis-(4-biphenyl)-1,3,4-oxadiazol:

p-Quaterphenyl-4-4'''-disulfonicacid Disodiumsalt:

p-Quaterphenyl-4-4'''-disulfonicacid Dipotassiumsalt:

4,4'''-Bis-(2-butyloctyloxy)-p-quanterphenyl:

3,5,3''',5''''-Tetra-t-butyl-p-sexiphenyl:

2-(1-Naphthyl)-5-phenyloxazol:

2-(4-Biphenyl)-6-phenylbenzoxazotetrasulfonicacid

Potassium Salt:

2-(4-Biphenyl)-6-phenylbenzoxazol-1,3:

4,4'-Diphenylstilbene:

[1,1'-Biphenyl]-4-sulfonic acid,

4,4"-1,2-ethene-diylbis-,dipotassium salt:

2,5-Bis-(4-biphenyl)-oxazol:

2,2'-([1,1'-Biphenyl]-4,4'-diyl-di-2,1-ethenediyl)-bis-benzenesulfonic acid Disodium Salt:

7-Amino-4-methylcarbostyryl:

1,4-Di[2-(5-phenyloxazolyl)]benzene:

7-Hydroxy-4-methylcoumarin:

p-Bis(o-methylstyryl)-benzene:

Benzofuran,2,2'-[1,1'-biphenyl]-4,4'-diyl-bis-tetrasulfonic acid:

7-Dimethylamino-4-methylquinolom-2:

7-Amino-4-methylcoumarin:

2-(p-Dimethylaminostyryl)-pyridylmethyl Iodide:

7-Diethylaminocoumarin:

7-Diethylamino-4-methylcoumarin:

2,3,5,6-1H,4H-Tetrahydro-8-methylquinolizino-[9,9a,1-gh]-coumarin:

7-Diethylamino-4-trifluormethylcoumarin:

7-Dimethylamino-4-trifluormethylcoumarin:

7-Amino-4-trifluormethylcoumarin:

2,3,5,6-1H,4H-Tetrahydroquinolizino-[9,9a,1-gh]-coumarin:

7-Ethylamino-6-methyl-4-trifluormethylcoumarin:

7-Ethylamino-4-trifluormethylcoumarin:

2,3,5,6-1H,4H-Tetrahydro-9-carboethoxyquinolizino-[9,9a,1-gh] coumarin:

2,3,5,6-1H,4H-Tetrahydro-9-(3-pyridyl)-quinolizino-[9,9a,1-gh] coumarin:

3-(2'-N-Methylbenzimidazolyl)-7-n,n-diethylaminocoumarin:

2,3,5,6-1H,4H-Tetrahydro-9-acetylquinolizino-[9,9a,1-gh]-coumarin:

N-Methyl-4-trifluormethylpiperidino-[3,2-g]-coumarin:

2-(p-Dimethylaminostyryl)-benzothiazolyethyl Iodide:

3-(2'-Benzimidazolyl)-7-N,N-diethylaminocoumarin:

Brillantsulfaflavin:

3-(2'-Benzothiazolyl)-7-diethylaminocoumarin:

2,3,5,6-1H,4H-Tetrahydro-8-trifluormethylquinolizino-[9,9a,1-gh] coumarin:

3,3'-Diethyloxacarbocyanine Iodide:

3,3'-Dimethyl-9-ethylthiacarbocyanine Iodide:

Disodium Fluorescein (Uranin):

9-(o-Carboxyphenyl)-2,7-dichloro-6-hydroxy-3H-xanthen-3-on2  
,7-Dichlorofluorescien • Fluorescein 548:

Fluorol 555 (Fluorol 7GA):

o-(6-Amino-3-imino-3H-xanthen-9-yl)-benzonic acid (Rhodamine  
560):

Benzoic Acid, 2-[6-(ethylamino)-3-(ethylimino)-2,7-dimethyl-3  
H-xanthen-9-yl], perchlorate (Rhodamine 575):

Benzonic

Acid, 2-[6-(ethylamino)-3-(ethylimino)-2,7-dimethyl-3X-xanth  
en-9-yl]-ethylester, monohydrochloride (Rhodamine 590):


1,3'-Diethyl-4,2'-quinolyloxacarbocyanine Iodide:

1,1'-Diethyl-2,2'-carbocyanine Iodid:

2-[6-(diethylamino)-3-(diethylimino)-3H-xanthen-9-yl]

benzonic acid (Rhodamine 610):

Ethanaminium, N-[(6-diethylamino)-9-(2,4-disulfophenyl)-3H-x  
anthen-3-ylidene]-N-ethylhydroxid, inner salt, sodium salt:

Malachit Green: 

3,3'-Diethylthiacarbocyanine Iodide:

1,3'-Diethyl-4,2'-quinolylthiacarbocyanine Iodide:

8-(2-Carboxyphenyl)-2,3,5,6,11,12,14,15-octahydro-1H,4H,10H  
,13H-diquinolizino[9,9a,1-bc:

9',9a',1-hi]xanthylum Perchlorate

(Rhodamine 640):

4-Dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran:

3,3'-Diethyloxadicarbocyanine Iodide:

8-(2,4-Disulfophenyl)-2,3,5,6,11,12,14,15-octahydro-1H,4H,10H,13H-diquinolizino [9,9a,1-bc:9',1-hi]xanthene

(Sulforhodamine 640):

5,9-Diaminobenzo[a]phenoxazonium Perchlorate:

9-Diethylamino-5H-benzo(a)phenoxazin-5-one:

5-Amino-9-diethylimino(a)phenoxazonium Perchlorate:

3-Ethylamino-7-ethylimino-2,8-dimethylphenoxazin-5-ium

Perchlorate:

8-(Trifluoromethyl)-2,3,5,6,11,12,14,15-octahydro-1H,4H,10H,13H-diquinolizino[9,9a,1-bc:9',9a,1-hi]xanthylum

Perchlorate:

1-Ethyl-2-(4-(p-Dimethylaminophenyl)-1,3-butadienyl)-pyridinium Perchlorate:

Carbazine 122:

9-Ethylamino-5-ethylimino-10-methyl-5H-benzo(a)phenoxazonium Perchlorate:

3-Diethylamino-7-diethyliminophenoxazonium Perchlorate:

3-Diethylthiadicarbocyanine Iodide:

Oxazine 750:

1-Ethyl-4-(4-(p-Dimethylaminophenyl)-1,3-butadienyl)-pyridinium Perchlorate:

1,1',3,3,3',3'-Hexamethylindodicarbocyanine Iodide:

1,1'-Diethyl-4,4'-carbocyanine Iodide:

2-(4-(p-Dimethylaminophenyl)-1,3-butadienyl)-1,3,3-trimethyl-3H-indolium Perchlorate:

2-(4-(p-Dimethylaminophenyl)-1,3-butadienyl)-3-ethylbenzothiazolium Perchlorate:

1,1'-Diethyl-2,2'-dicarbocyanine Iodide:

1-Ethyl-4-(4-(9-(2,3,6,7-tetrahydro-1H,5H-benzo(i,j)-chinolizinium))-1,3-butadienyl)-pyridinium Perchlorate:

3,3'-Dimethyloxatricarbocyanine Iodide:

1-Ethyl-4-(p-Dimethylaminophenyl)-1,3-butadienyl)-quinolinium Perchlorate:

8-Cyano-2,3,5,6,11,12,14,15-octahydro-1H,4H,10H,13H-diquinolizino[9,9a,1-bc:9a',1-hi]xanthylum Perchlorate (Rhodamine800):

2-(6-(4-Dimethylaminophenyl)-2,4-neopentylene-1,3,5)-3-methylbenzothiazolium Perchlorate:

1,1',3,3,3',3'-Hexamethylindotricarbocyanine Iodide:

IR125 :

3,3'-Diethylthiatricarbocyanine Iodide :

IR144 :

2-(6-(9-(2,3,6,7-Tetrahydro-1H,5H-benzo(i,j)-chinolizinium))-2,4-neopentylene-1,3,5-hexatrienyl)-3-methylbenzothiazolium Perchlorate:

3,3'-Diethyl-9,11-neopentylene-thiatricarbocyanine Iodide:

1,1',3,3,3',3'-Hexamethyl-4,4',5,5'-dibenzo-2,2'-indotricarbocyanine Iodide:

3,3'-Diethyl-4,4',5,5'-dibenzothiatricarbocyanine Iodide:

1,2'-Diethyl-4,4'-dicarbocyanine Iodide:

IR140:

2-(8-(4-p-Dimethylaminophenyl)-2,4-neopentylene-1,3,5,7-octatetraenyl)-3-methylbenzothiazolium Perchlorate:

IR132 :

2-(8-(9-(2,3,6,7-Tetrahydro-1H,5H-benzo(i,j)chinolizinium))-2,4-neopentylene-1,3,5,7-octatetraenyl)-3-methylbenzothiazolium Perchlorate:

IR26:

IR 5.

40. The solid dye laser according to any one of claims 35 to 39, wherein a pulse laser beam by the short pulse laser has a pulse width of less than 10 nsec.

41. The solid dye laser according to any one of claims 35 to 40, being capable to exchange the solid laser medium without changing an optical system.

42. The solid dye laser according to any one of claims 35 to 41, further comprising an optical grating for controlling a laser oscillation wavelength.

43. The solid dye laser according to claim 42, being capable to scan a wavelength by controlling spatial positions of an excitation light and a prism.

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44. The double-resonance-absorption microscope according to any one of claims 1 to 34, wherein the solid dye laser according to any one of claims 35 to 43 is provided as either or both of the light source for the pump light and the light source for the erase light.

45. A double-resonance-absorption microscope comprising a light source for a pump light which excites a sample molecule to a first electronic excited state from a ground state and a light source for an erase light which excites the sample molecule to a second electronic excited state or a higher excited state from the first electronic excited state, wherein a light emission upon deexcitation of the sample molecule to the ground state from each of the electronic excited state is detected,

said double-resonance-absorption microscope characterized in that the pump light has a photon energy less than  $1/2$  of an excitation energy which excites the sample molecule to the first electronic excited state from the ground state.

46. A double-resonance-absorption microscope comprising a light source for a pump light which excites a sample molecule to a first electronic excited state from a ground state and a light source for an erase light which excites the sample molecule to a second electronic excited state or a higher excited state from the first electronic excited state, wherein a light emission upon deexcitation of the sample molecule to the ground state is detected from each of the electronic excited state,



said double-resonance-absorption microscope characterized in that the erase light has a photon energy less than  $1/2$  of an excitation energy which excites the sample molecule to the second electronic excited state from the first electronic excited state.

47. A double-resonance-absorption microscope comprising a light source for a pump light which excites a sample molecule to a first electronic excited state from a ground state and a light source for an erase light which excites the sample molecule to a second electronic excited state or a higher excited state from the first electronic excited state and wherein a light emission upon deexcitation of the sample molecule to the ground state from each of the electronic excited state is detected, said double-resonance-absorption microscope characterized in that the pump light has a photon energy less than  $1/2$  of an excitation energy which excites the sample molecule to the first electronic excited state from the ground state and the erase light has a photon energy less than  $1/2$  of an excitation energy which excites the sample molecule to the second electronic excited state from the first ground state.

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48. The double-resonance-absorption microscope according to any one of claims 45 to 47, wherein a photon energy range of a light emission from the sample molecule in the first electronic excited state and a excitation energy range which excites the sample molecule to the second electronic excited state from the

first electronic excited state overlap with each other.

49. The double-resonance-absorption microscope according to any one of claims 45 to 48, further comprising overlap means for partially overlapping irradiating areas of the pump light and the erase light with each other, wherein an emission area upon deexcitation of the sample molecule to the ground state from the first electronic excited state is partially inhibited by irradiating the pump light and the erase light through the overlap means.

50. The double-resonance-absorption microscope according to any one of claims 45 to 49, wherein a sample is dyed with a fluorescence labeler molecule having at least three electronic state including a ground state and the sample molecule is the ~~fluorescence labeler molecule.~~

51. A double-resonance-absorption microscope comprising:  
a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state of a singlet state from a ground state;

a light source for a probe light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher excited state of a singlet state from the first electronic excited state; and

overlap means for overlapping a part or all of irradiating areas of the pump light and the probe light with each other;  
wherein a sample is irradiated with the pump light and

the probe light through the overlap means, and a transient raman scattering light emitted from an area in the sample where the pump light and the probe light overlap with each other is detected.

52. A double-resonance-absorption microscope comprising:

a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state of a singlet state from a ground state;

a light source for a probe light of a wavelength  $\lambda_3$  which excites the sample molecule, transited to a triplet level lower in energy than the first electronic excited state from the first electronic excited state, to a higher excited triplet level from the triplet level; and

overlap means for overlapping a part or all of irradiating areas of the pump light and the probe light with each other;

wherein a sample is irradiated with the pump light and the probe light through the overlap means, and a transient raman scattering light emitted from an area in the sample where the pump light and the probe light overlap with each other is detected.

53. A double-resonance-absorption microscope comprising:

a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state of a singlet state from a ground state;

a light source for a probe light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher excited state of a singlet state from the first

electronic excited state;

a light source for a probe light of a wavelength  $\lambda_3$  which excites the sample molecule, transited to a triplet level lower in energy than the first electronic excited state from the first electronic excited state, to a higher excited triplet level from the triplet level;

overlap means for overlapping a part or all of irradiating areas of the pump light and the probe light with each other; and

irradiation time control means for controlling the time to irradiate a sample with the probe light of the wavelength  $\lambda_2$  and the probe light of the wavelength  $\lambda_3$ ;

wherein the sample is irradiated with the pump light and the probe light through the overlap means, and by the irradiation time control means, the probe light of the wavelength  $\lambda_2$  is applied to the sample before the sample molecule transits to the triplet level from the first electronic excited state and the probe light of the wavelength  $\lambda_3$  is applied to the sample after the sample molecule transits to the triplet level from the first electronic state, and a transient raman scattering light emitted from an area in the sample where the pump light and the probe light overlap with each other is detected.

54. The double-resonance-absorption microscope according to claim 53 wherein the irradiation time control means is a delay optical system which controls a time to irradiate the sample

by controlling an optical path of the probe light.

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55. The double-resonance-absorption microscope according to any one of claims 51 to 54, further comprising means for adjusting a polarization state or a wavelength or a strength of the pump light and the erase light.

56. The double-resonance-absorption microscope according to any one of claims 51 to 55 wherein the sample molecule to be excited is a cation.

57. A fluorescence correlation method comprising the steps of:  
condensing a pulse excitation light onto a sample;  
operating a position of the pulse excitation light relatively to the sample;  
measuring a fluorescence correlation function at each operation position; and  
calculating the fluorescence correlation function where a signal component of a measuring time region having no fluorescence emission is excluded.

58. The fluorescence correlation function according to claim 57 used with a double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state from a ground state, a light source for a erase light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher electronic excited state

from the first electronic excited state and overlap means for partially overlapping irradiating areas of the pump light and the erase light with each other, wherein an emission area upon deexcitation of the sample to the ground state from the first electronic state is partially inhibited by irradiating the pump light and the erase light through the overlap means.

59. The fluorescence correlation function according to claim 57 used with a double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state from a ground state, a light source for a erase light of a wavelength  $\lambda_2$  which excites the sample molecule to a second electronic excited state or a higher electronic excited state from the first electronic excited state and overlap means for partially overlapping irradiating areas of the pump light and the erase light with each other, wherein a sample is irradiating with the pump light and the erase light through the overlap means, and a photoresponse from an area in the sample where the pump light and the erase light overlap with each other is detected.

60. The fluorescence correlation method according to claim 57 used with a double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state of a singlet state from a ground state, a light source for a probe light of a wavelength  $\lambda_2$  which excites the sample

molecule to a second electronic excited state or a higher excited state of a singlet state from the first electronic excited state and overlap means for overlapping a part or all of irradiating areas of the pump light and the probe light with each other, wherein a sample is irradiated with the pump light and the probe light through the overlap means, and a transient raman scattering light emitted from an area in the sample where the pump light and the probe light overlap with each other is detected.

61. The fluorescence correlation method according to claim 57 used with a double-resonance-absorption microscope comprising a light source for a pump light of a wavelength  $\lambda_1$  which excites a sample molecule to a first electronic excited state of a singlet state from a ground state, a light source for a probe light of a wavelength  $\lambda_3$  which excites the sample molecule, transited to a triplet level lower in energy than the first electronic excited state from the first electronic excited state, to a higher excited triplet level from the triplet level and overlap means for overlapping a part or all of irradiating areas of the pump light and the probe light with each other, wherein a sample is irradiated with the pump light and the probe light through the overlap means, and a transient raman scattering light emitted from an area in the sample where the pump light and the probe light overlap with each other is detected.

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